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14. ABSTRACT

Funds were requested for the purchase of an ultrafast laser system and optical parametric amplifier (OPA) for nonlinear optical diagnostics in flames at data rates of 5 kHz. In the time period between the submission of the DURIP proposal and the initiation of the grant, the specifications for commercial ultrafast laser systems in the price range of interest had considerably improved. A Coherent laser system with pulse energies of 2.6 mJ at 5 kHz and 1.0 mJ at 10 kHz repetition rate was purchased. The ultrafast laser system includes at 128-pixel pulse shaper to correct the phase of the laser spectrum and to ensure that the amplified laser pulses are very close to Fourier-transform-limited. An OPA and frequency-mixing system were purchased to produce a tunable beam that will be used for the pump radiation in the femtosecond coherent anti-Stokes Raman scattering (CARS) measurements. This system has been delivered and installed in the Prof. Lucht's Applied Spectroscopy Laboratory.

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(DURIP 09)- ULTRAFAST LASER SYSTEM FOR COHERENT ANTI-STOKES RAMAN SCATTERING MEASUREMENTS AT DATA RATES OF 5 KHZ

AFOSR Grant Number: FA9550-09-1-0387

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Executive Summary

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Project Abstract:

Funds were requested for the purchase of an ultrafast laser system and optical parametric amplifier (OPA) for nonlinear optical diagnostics in flames at data rates of 5 kHz. The ultrafast laser system as requested in the proposal system operated at 5 kHz with a pulse energy of >1.2 mJ at the fundamental wavelength of approximately 800 nm. In the time period between the submission of the DURIP proposal and the initiation of the grant, the specifications for commercial ultrafast laser systems in the price range of interest had considerably improved. A Coherent laser system with pulse energies of 2.6 mJ at 5 kHz and 1.0 mJ at 10 kHz repetition rate was purchased. The ultrafast laser system includes at 128-pixel pulse shaper to correct the phase of the laser spectrum and to ensure that the amplified laser pulses are very close to Fourier-transform-limited. An OPA and frequency-mixing system were purchased to produce a tunable beam that will be used for the pump radiation in the femtosecond coherent anti-Stokes Raman scattering (CARS) measurements. This system has been delivered and installed in the Prof. Lucht's Applied Spectroscopy Laboratory.

The Coherent ultrafast laser system will be used for high-data-rate, single-pulse coherent anti-Stokes Raman scattering measurements. In addition, the use of the system for two-photon-induced fluorescence detection and measurement of species such as OH, CH, and NO will be explored. The potential for planar imaging of two-photon-induced fluorescence from these species at data rates of up to 5 kHz will be explored.

AFOSR grant funds for the project have been fully (100%) expended.

Project Objectives

The objective of the DURIP grant was to purchase an ultrafast laser system to develop and demonstrate advanced laser diagnostic techniques. In particular, the ultrafast laser system was selected to advance the state-of-the-art with respect to femtosecond (fs) coherent anti-Stokes Raman scattering (CARS) spectroscopy. Fs CARS offers some significant potential advantages compared with nanosecond (ns) CARS, i.e., CARS as usually performed with ns pump and Stokes lasers. These potential advantages include (1) performing real-time temperature and species measurements at data rates of 1 kHz or greater, (2) the absence of any effect of collisions in the determination of temperature and concentration from the fs CARS signal, and (3) higher signal-to-noise ratios due to the nearly Fourier-transform-limited (FTL) nature of the ultrafast laser radiation. In collaboration with Sukesh Roy at Spectral Energies and James Gord at AFRL, the Lucht group has been developing fs CARS techniques for temperature and concentration measurements (Lucht et al., 2006; Lucht et al., 2007; Roy et al. 2008; Roy et al., 2009a; Roy et al., 2009b; Richardson et al., 2010). The experiments described in these papers were all performed at Wright-Patterson Air Force Base using a Coherent ultrafast laser system with a repetition rate of 1 kHz and a pulse energy of approximately 1 mJ in the fundamental beam at 800 nm. While these initial experiments were very promising, extension of the fs CARS techniques to data rates of 5 kHz and higher is highly desirable so that a greater range of turbulence frequencies can be measured. An ultrafast laser system with repetition rates up to 10 kHz and a fundamental beam pulse energy of 1 mJ even at this very high repetition rate was therefore purchased.

Coherent Ultrafast Laser System

The Coherent ultrafast laser system that was purchased with the DURIP funds is schematically illustrated in Fig. 1. The mode-locked oscillator for the system is a Mantis. The Mantis has a repetition rate of 80 MHz, an average power of > 500 mW, and a bandwidth of greater than 70 nm; the bandwidth is sufficient to produce pulses as short as 30 fs. The Mantis operates at a fixed wavelength of 800 nm. An optically pumped semiconductor laser (OPSL) is incorporated in the Mantis box and serves as the pump laser for the titanium-sapphire (Ti:S) slab crystal in the mode-locked oscillator cavity.

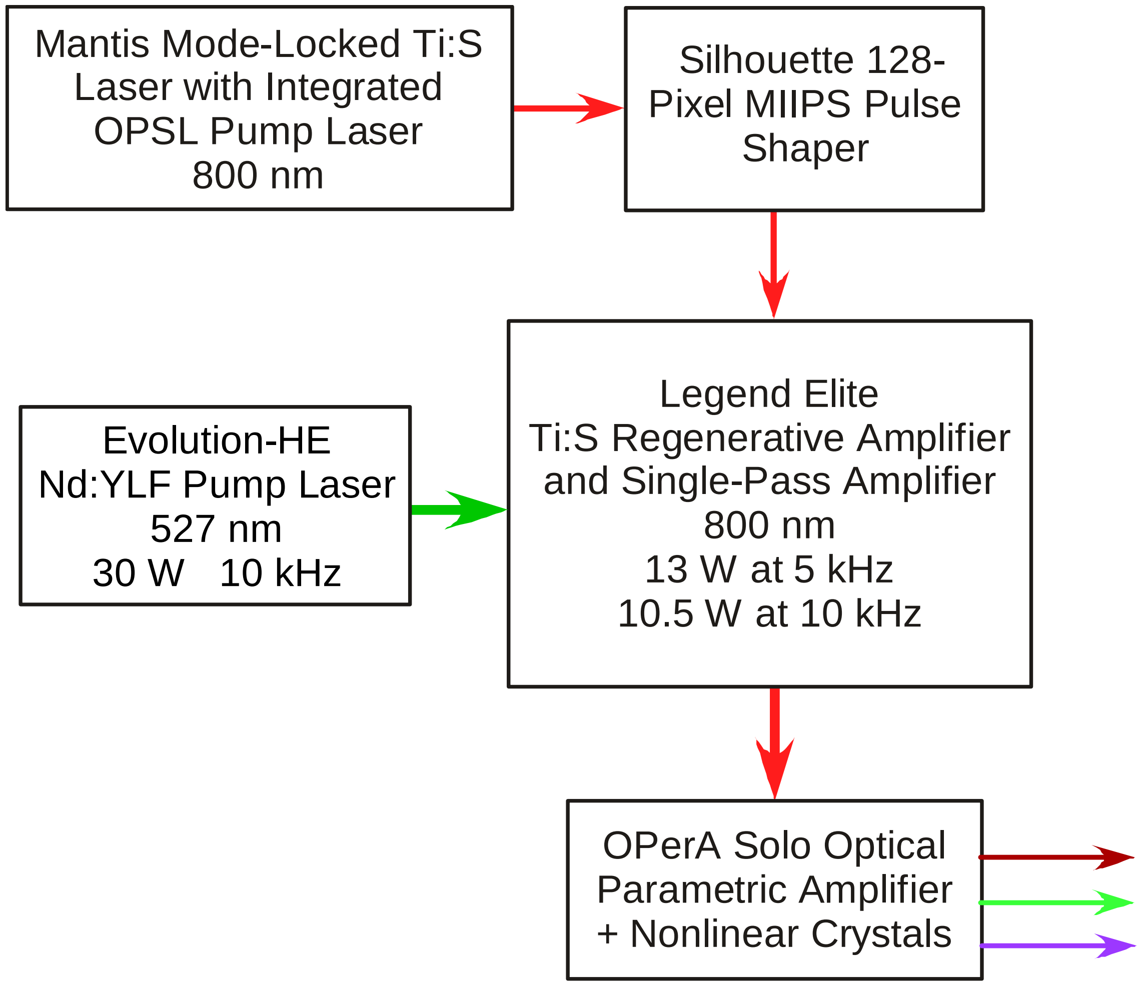

Fig. 1. Schematic diagram of the Coherent ultrafast laser system that was purchased with DURIP funds.

The Mantis beam is then directed into the Silhouette pulse shaper/pulse characterizer unit. The Silhouette has a 128-pixel liquid crystal spatial light modulator. The pulse can be characterized using the technique of multiphoton intrapulse interference phase scan (MIIPS) with the Silhouette. The Silhouette is used to correct the phase distortions that cause a departure from the Fourier transform limit.

The pump laser for the Legend Elite amplifier is an Evolution HE Nd:YLF laser that is diode-pumped and intracavity-frequency double to produce a 527 nm output beam. At 5 kHz 70 W of average power are used to pump the amplifier, at 10 kHz 75 W of average power is used to pump the amplifier.

The Legend Elite ultrafast amplifier is used to amplify pulses from the Mantis. The Legend Elite system includes both a regenerative amplifier stage and a single-pass amplifier stage. In both of these stages the slab Ti:S crystals are mounted on thermoelectric coolers. The Legend Elite amplifier system is capable of operation at repetition rates of either 5 kHz or 10 kHz. It also includes stretchers and compressors for either 30 fs or 60 fs pulses. During installation all four repetition rate/pulse length combinations were demonstrated. The original specification for the system pulse length were 40 fs and 90 fs. However, with the incorporation of the Silhouette into the system the actual measured pulses were 30 fs and 60 fs instead of 40 fs and 90 fs, respectively. The shorter pulse lengths were achieved because the Silhouette corrects the phase of the spectrum of pulses at the input to the amplifier so that the output pulses are within 1-2% of the Fourier transform limit. Consequently the shortest possible pulses can be extracted from the amplifier.

The output of the amplifier is then directed into the OPerA Solo, an automated optical parametric amplifier (OPA) with a harmonic generation stage after the OPA. The OPA is used to produce signal and idler pulses from the input pump pulse at 800 nm. The OPA is seeded with a white light continuum. The signal wavelengths from the OPA process range from 1140 nm to 1600 nm and the idler wavelengths range from 2600 nm to 1600 nm. The crystal for second harmonic generation of the signal beam was purchased and installed in the OPerA Solo. The wavelength range of the frequency-doubled signal beam is from 580 nm to 800 nm. For the planned fs CARS measurements, the pump beam wavelengths, powers, and pulse energies for different Raman species are listed in Table 1. For all of the species listed, the pump pulse energy is considerably higher than the pump pulse energy of 30 μJ for the single-pulse measurements reported by Roy et al. (2009b). Consequently it is anticipated that the signal to noise ratios for the planned fs CARS measurements will be considerably improved compared to the already excellent results reported by Roy et al. (2009b).

Summary

The ultrafast laser system requested in the DURIP proposal has been purchased and installed. The specifications and characteristics of the laser system that was actually purchased are superior to the system that was specified in the proposal due to rapid advances in the technology of commercial ultrafast laser systems. The Coherent ultrafast laser system will be

used first for fs CARS measurements, but should be very useful for a wide range of nonlinear diagnostic methods for reacting flows.

Table 1. Pump wavelength, average power, and pulse energy for different species of interest. The Stokes wavelength is assumed to be 800 nm.

Species	Raman Shift (cm^{-1})	Pump λ (nm)	Pump Ave. Power (W), 5 kHz	Pump Pulse Energy (μJ), 5 kHz
CO_2	1388	720	630	126
O_2	1556	711	650	130
N_2	2330	674	735	147
CH_4	2915	649	690	138
H_2O	3657	619	585	117
H_2	4160	600	425	85

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